

Processing and Deposition of Nanocrystalline Oxide Composites for  
Thermal Barrier Coatings

Technical Report on ONR Grant No. N00014-95-1-0626  
for the period of April 1, 2000-June 30, 2000

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***1. Electrophoretic Deposition of NiCrAlY Bond Coat and Oxide Nanocomposite***

As described in a previous report [1], electrophoretic deposition was used as the method in coating substrate materials for thermal barrier coating applications. In this report, we used electrophoretic deposition to deposit a NiCrAlY bond coat, and a nanocomposite  $\text{Al}_2\text{O}_3$ -YSZ coating, onto a Ni substrate. The bond coat forms a protective oxide scale that inhibits oxidation of the alloy substrate. The NiCrAlY interlayer can also provide good diffusion stability between the metallic substrate and the ceramic coating layer. We examined the thermal stability of the  $\text{Al}_2\text{O}_3$ -YSZ coatings when applied on top of the NiCrAlY-based bond coat. In particular, the effect of alumina content on the thermal stability of the nanocomposite coatings was examined.

For deposition of the bond coat, a coating solution consisting of ethanol, water, nitric acid, and the NiCrAlY powder was used. The coating composition was similar to that reported previously [1]. NiCrAlY powder was first mechanically milled for 24 hr to decrease particle size and then added to the coating solution to form a slurry. The slurry was then ultrasonicated for 15 min and aged for 24 hr. Ni electrodes were then lowered into the slurry and a DC voltage ( $\sim 40$  V/cm) was applied for 120 sec, resulting in a  $\sim 50$   $\mu\text{m}$ -thick coating of the cathode with the NiCrAlY powder. The electrodes were then removed from the slurry, and the coated Ni electrode was dried at  $110^\circ\text{C}$  for 2 hr.

Next,  $\text{Al}_2\text{O}_3$ -YSZ nanocomposites were coated onto NiCrAlY-coated substrates using electrophoretic deposition. A DC voltage ( $\sim 40$  V/cm) was applied for 300 sec, resulting in a  $\sim 50$   $\mu\text{m}$ -thick  $\text{Al}_2\text{O}_3$ -YSZ coating on top of the NiCrAlY bond coat. Following drying, the coatings were cold isostatically pressed at 50,000 psi to increase mechanical stability.

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## ***2. Thermal Stability of Alumina-Zirconia Thermal Barrier Coatings***

Oxide nanocomposite coatings with various  $\text{Al}_2\text{O}_3$  contents were prepared and tested for thermal stability in the presence and absence of a bond coat (Table 1). Following electrophoretic deposition, the  $\text{Al}_2\text{O}_3$ -YSZ/NiCrAlY-coated Ni substrates were pretreated in argon at 1150 °C for 1 hr (ramp = 10 °C/min). They were then tested for long-term thermal stability by heating to 1150 °C in air (ramp = 10 °C/min). In addition, the coatings were also subjected to thermal cycles of 25 °C-1150 °C-25 °C (with ramping and cooling rates of ~ 300 °C/min) to examine any failure modes due to thermal shock. The samples were inspected visually every hour for spallation of the coating and oxidation of the Ni substrate. All experiments were repeated three times.

The addition of the bond coat did not significantly increase the thermal stability of the thermal barrier coatings. Coatings prepared with and without the intermediate bond coat layer had similar thermal stability, in both coating lifetime and thermal cycling studies (Tables 1 and 2).

The addition of a small amount of  $\text{Al}_2\text{O}_3$  to the coatings had a positive effect on the coating lifetime in the presence and absence of a bond coat (Table 1). The coating without  $\text{Al}_2\text{O}_3$  began to crack and spall after 3-4 hr of heat treatment. The most stable compositions were 5 wt%  $\text{Al}_2\text{O}_3$ -95 wt% YSZ and 10 wt%  $\text{Al}_2\text{O}_3$ -90 wt% YSZ, which lasted for 6 hr before significant spallation occurred. Coatings with higher  $\text{Al}_2\text{O}_3$  contents (20 wt% and 100 wt%) had poor thermal stability, lasting only 2 hr and 1 hr, respectively, at 1150 °C before spallation occurred.

The reduced thermal stability when using > 10 wt%  $\text{Al}_2\text{O}_3$  could be attributed to a difference in the thermal expansion coefficients of  $\text{Al}_2\text{O}_3$  and YSZ, which could induce stress and subsequent failure of the coating. Thermal stabilization with the addition of 5-10 wt%  $\text{Al}_2\text{O}_3$  could be explained by a decrease in oxygen conductivity [2].

For the thermal cycling studies,  $\text{Al}_2\text{O}_3$  addition also had a positive effect on the stability of the coatings with and without the NiCrAlY bond coat (Table 2). The most stable compositions were 5 wt%  $\text{Al}_2\text{O}_3$ -95 wt% YSZ and 10 wt%  $\text{Al}_2\text{O}_3$ -90 wt% YSZ, which lasted 3 thermal cycles before significant spallation occurred. Other  $\text{Al}_2\text{O}_3$  compositions lasted  $\leq 1$  thermal cycle before failure.

Coatings were examined with X-ray diffraction (XRD) to determine their crystal phase before and after the thermal shock treatment. Figure 1 shows the XRD patterns of 5 wt%  $\text{Al}_2\text{O}_3$ -95 wt% YSZ before and after 3 thermal cycles at 1150 °C. Prior to the thermal cycling,  $\text{ZrO}_2$  was present in the tetragonal phase. Ni peaks were noted from the Ni substrate, after the powder was deposited onto the Ni substrate. After 3 thermal cycles,  $\text{ZrO}_2$  retained 100% tetragonal phase. NiO peaks were observed after the heat treatment, due to cracking of the coating and oxidation of the Ni substrate. Other  $\text{Al}_2\text{O}_3$ -YSZ nanocomposite coatings had similar phases present as this system before and after thermal shock treatment at 1150 °C.

### **3. Future Work**

#### *Plasma Spraying of NiCrAlY Bond Coat and Coating of the Oxide Nanocomposites*

NiCrAlY bond coats will be applied onto the Ni substrates using plasma spraying. Following the deposition of the plasma-sprayed NiCrAlY coatings, electrophoretic deposition will be used to deposit oxide nanocomposite coatings with various  $\text{Al}_2\text{O}_3$  contents. The thermal barrier coatings will be subjected to several pretreatments prior to coating lifetime and thermal shock studies. Following thermal stability studies, the coatings will be examined with XRD and SEM to determine the failure mechanisms responsible for the spallation of the oxide composite and NiCrAlY bond coat. The effect of  $\text{Al}_2\text{O}_3$  in coating stability will also be investigated.

### **4. References**

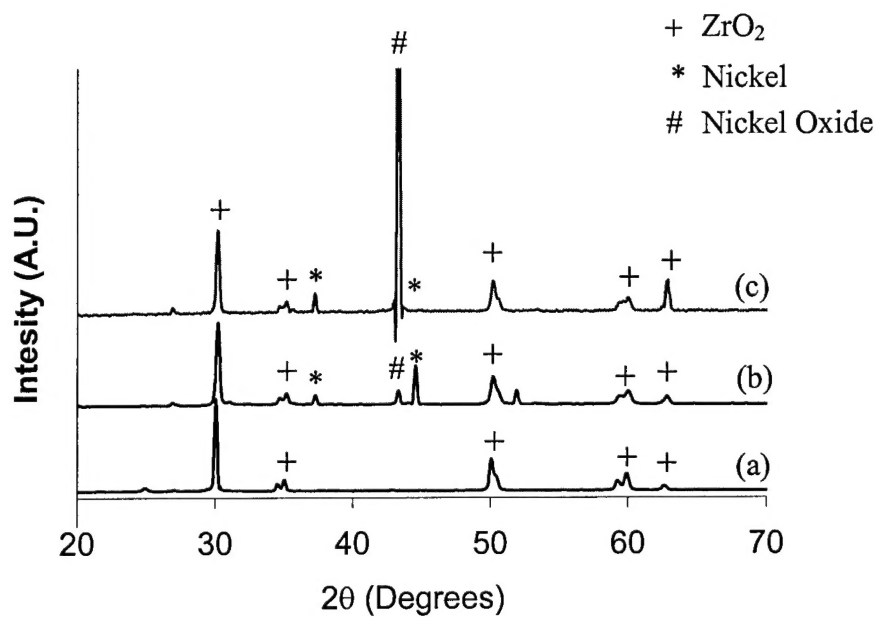
- [1] Ying, J.Y., Technical Report on ONR Grant No. N00014-95-1-0626 for the period of October 1, 1999-December 31, 1999.
- [2] Ying, J.Y., Technical Report on ONR Grant No. N00014-95-1-0626 for the period of January 1, 2000-March 31, 2000.

**Table 1.** Coating lifetime for various alumina-zirconia nanocomposite coatings. The coating lifetime was given by the duration before spallation was first observed for a coating heat treated at 1150 °C in air.

Coating Composition	Coating lifetime without bond coat (hr)	Coating lifetime with bond coat (hr)
100 wt% YSZ	3	4
5 wt% Al <sub>2</sub> O <sub>3</sub> -95 wt% YSZ	6	6
10 wt% Al <sub>2</sub> O <sub>3</sub> -90 wt% YSZ	6	6
20 wt% Al <sub>2</sub> O <sub>3</sub> -80 wt% YSZ	1	2
100 wt% Al <sub>2</sub> O <sub>3</sub>	1	1

**Table 2.** Thermal shock stability for various alumina-zirconia nanocomposite coatings. The number of cycles to failure was given by the number of thermal cycles prior to the observation of spallation. A thermal cycle involved heating the sample in air from 25 °C to 1150 °C, holding at 1150 °C for one hour, and then cooling to 25 °C, with ramping and cooling rates of ~ 300 °C/min.

Coating Composition	Cycles to failure without bond coat (hr)	Cycles to failure with bond coat (hr)
100 wt% YSZ	1	1
5 wt% Al <sub>2</sub> O <sub>3</sub> -95 wt% YSZ	3	3
10 wt% Al <sub>2</sub> O <sub>3</sub> -90 wt% YSZ	3	3
20 wt% Al <sub>2</sub> O <sub>3</sub> -80 wt% YSZ	1	1
100 wt% Al <sub>2</sub> O <sub>3</sub>	<1	<1



**Figure 1.** X-ray diffraction patterns of 5 wt%  $\text{Al}_2\text{O}_3$ -95 wt% YSZ (a) powder, (b) as deposited on Ni substrate with NiCrAlY bond coat, and (c) following 3 thermal cycles at 1150 °C.

# REPORT DOCUMENTATION PAGE

Form Approved  
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1. AGENCY USE ONLY (Leave Blank)	2. REPORT DATE 1 July, 2000	3. REPORT TYPE AND DATES COVERED Progress Report: 1 April 2000 - 31 June 2000
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4. TITLE AND SUBTITLE Processing and Deposition of Nanocrystalline Oxide Composites for Thermal Barrier Coatings	5. FUNDING NUMBERS G - N00014-95-1-0626
6. AUTHORS Jackie Y. Ying Justin T. McCue	

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Department of Chemical Engineering Massachusetts Institute of Technology 77 Massachusetts Avenue, Room 66-544 Cambridge, MA 02139-4307	8. PERFORMING ORGANIZATION REPORT NUMBER
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9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Office of Naval Research 800 North Quincy Street Ballston Tower One Arlington, VA 22217-5660	10. SPONSORING / MONITORING AGENCY REPORT NUMBER
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11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION / AVAILABILITY STATEMENT  Approved for pulbic release; distribution unlimited	12b. DISTRIBUTION CODE
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13. ABSTRACT (Maximum 200 words)  
This report describes the thermal stability of nanocrystalline oxide composites for thermal barrier coating applications. The coatings were tested fo long-term and thermal shock stability. Electrophoretic deposition (EPD) was used to coat the nanocomposite powders onto nickel substrates. A NiCrAlY bond coat was also coated onto the Ni substrates prior to deposition of the nanocrystalline oxide composites. It was found that a small addition of alumina (5 wt%) enhanced the thermal stability of the coatings. The NiCrAlY bond coat, when coated using electrophoretic deposition did not significantly enhance the thermal stability of the coatings. Future work will combine plasma-sprayed bond coats with oxide composites deposited via EPD.

14. SUBJECT TERMS Nanocrystalline, Electrophoretic Deposition, Thermal Barrier Coatings	15. NUMBER OF PAGES 5
	16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL
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